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Proceedings of the XXIII International School of Semiconducting Compounds, Jaszowiec 1994

IS THE MAGNETISM OF HgSe:Fe CLEARLY UNDERSTOOD?*

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The high magnetic field magnetization measurements of low composition $\text{Hg}_{1-x}\text{Fe}_x\text{Se}$ ($0.0002 < x < 0.003$) at temperatures ranging from 1.6 K to 40 K and in magnetic fields up to 20 T are reported. The magnetization in a sample containing only Fe^{3+} ions ($x = 0.0002$) is described by the Brillouin function ($j = 5/2$). The magnetization in the sample codoped with Ga, containing only Fe^{2+} is well described in the model of isolated Fe^{2+} ions. The latter model takes into account the tetrahedral crystal field, the spin-orbit interaction and the Zeeman term. On the other hand, the magnetization of sample with both Fe^{3+} and Fe^{2+} present simultaneously ($x = 0.003$) is shown to be different from a simple additive superposition of the contributions due to Fe^{3+} and Fe^{2+} subsystems. The possible origin of this discrepancy is discussed.

PACS numbers: 75.20.Hr, 75.30.Hx

The high magnetic field magnetization data for diluted magnetic semiconductors (DMS) in which a fraction of cations were substituted by Fe ions [1] are available up to date for $\text{Cd}_{1-x}\text{Fe}_x\text{Se}$ [2], $\text{Cd}_{1-x}\text{Fe}_x\text{Te}$ [3], $\text{Zn}_{1-x}\text{Fe}_x\text{Se}$ [4], $\text{Hg}_{1-x}\text{Fe}_x\text{Se}$ [5]. The studies presented in [5] covered the composition range $0.003 < x < 0.05$ and mainly dealt with the anisotropy of magnetic interactions. The aim of the present paper is to investigate the high field magnetization of $\text{Hg}_{1-x}\text{Fe}_x\text{Se}$ in very dilute limit of composition ($0.0002 < x < 0.003$). In this case two qualitatively different regimes of composition x can be distinguished: (i) for smallest compositions ($x < 0.0003$) all iron impurities are ionized to the $\text{Fe}^{3+}(3d^5)$ state with one electron donated to the conduction band, (ii) for composition $x > 0.0003$, while $4.5 \times 10^{18} \text{ cm}^{-3}$ irons are in $\text{Fe}^{3+}(3d^5)$ state, the remaining part is in $\text{Fe}^{2+}(3d^6)$ state. Thus, in regime (ii) two types of iron ions coexist in the crystal. The reported EPR measurements [6] for samples with $0.0006 < x < 0.003$ unambiguously

*Work partially supported by U.S.-Poland Maria Curie-Skłodowska Joint Fund II under grant No. PAN/NSF-92-113.

confirmed the existence of $\text{Fe}^{3+}(3d^5)$ in the material. We studied samples coming from these two regimes. We also studied a sample with $x = 0.003$ codoped with Ga, which ensured that iron existed *only* in the Fe^{2+} state.

Magnetic behaviour of $\text{Fe}^{3+}(3d^5)$ ions should not differ from that of a well known $\text{Mn}^{2+}(3d^5)$ ions given by spin moment only $s = 5/2$, while the orbital moment $l = 0$. The ground state of such ions is spin sextet. Consequently, this should lead to the Brillouin-type paramagnetic behaviour of the magnetization. On the other hand, the $\text{Fe}^{2+}(3d^6)$ ions possess apart from the spin moment also the orbital moment ($s = 2$, $l = 2$). The model calculations [7] of isolated $\text{Fe}^{2+}(3d^6)$ ion in a tetrahedral environment showed that the ground state of Fe^{2+} is magnetically inactive singlet. That leads to the field induced magnetic moment only, known as Van Vleck paramagnetism. It is characterized by temperature independent susceptibility at low temperatures and a very weak saturation of the magnetization in high magnetic fields. Such description of $\text{Fe}^{2+}(3d^6)$ in II-VI compounds with some modifications (additional trigonal distortion) to account for magnetization anisotropy was successfully applied to CdTe, CdSe, ZnSe [1-4]. The presented magnetization measurements of HgSe:Fe were performed at temperatures from

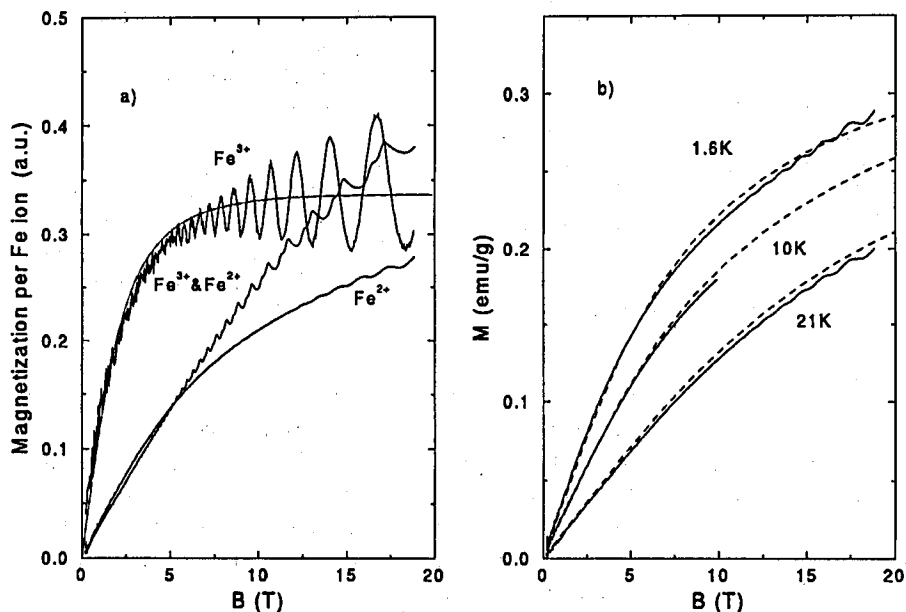


Fig. 1. (a) Magnetization of $\text{Hg}_{1-x}\text{Fe}_x\text{Se}$ per Fe ion at temperature 4.2 K for three samples labelled as follows: Fe^{3+} — $x = 0.0002$, Fe^{2+} and Fe^{3+} — $x = 0.003$, Fe^{2+} — $x = 0.003$: $7 \times 10^{18} \text{ cm}^{-3}$ Ga. Solid lines — experiment, dashed line — Brillouin function $j = 5/2$; (b) magnetization of $\text{Hg}_{0.997}\text{Fe}_{0.003}\text{Se}$: $7 \times 10^{18} \text{ cm}^{-3}$ Ga at 1.6, 10, 21 K. Solid lines — experiment, dashed lines — theory.

1.6 K to 40 K in the magnetic fields up to 20 T. The high precision capacitive torsional magnetometer was used to collect the magnetization data continuously (approximately 1000 points per 20 T) during the sweep of the magnet. The results of the magnetization measurements for three samples at 4.2 K are shown in Fig. 1a. The data presented in this figure were corrected for diamagnetic lattice susceptibility of the host material — HgSe. The magnetization was normalized to one Fe atom. The oscillatory behaviour superimposed on the magnetization of Fe ions into high magnetic fields originates in the magnetization of free electrons. Because of relatively low Fe compositions, this contribution is clearly visible. The sample with $x = 0.0002$ exhibits classical Brillouin type behaviour described by Brillouin function with $j = 5/2$ (represented by dashed line in Fig. 1a). The above result confirms the similarity of $\text{Fe}^{3+}(3d^5)$ and $\text{Mn}^{2+}(3d^5)$ ions and is, in fact, the first measurement of the magnetization on such very dilute sample HgSe:Fe. The result for the next sample with $x = 0.003$ codoped with $7 \times 10^{18} \text{ cm}^{-3}$ Ga is represented by lowest trace in Fig. 1a. This sample contains only $\text{Fe}^{2+}(3d^6)$ ions. The character of this dependence on magnetic field is satisfactorily reproduced in the model assuming the presence of $\text{Fe}^{2+}(3d^6)$ ions with the Hamiltonian including the tetrahedral crystal field, spin-orbit interaction and the Zeeman term. The result of such calculations is presented in Fig. 1b for several temperatures. We took crystal field splitting $10Dq = 2430 \text{ cm}^{-1}$ and spin-orbit interaction $\lambda = 106 \text{ cm}^{-1}$. The most interesting, and qualitatively different, is the magnetization of the sample with $x = 0.003$, when $\text{Fe}^{3+}(3d^5)$ and $\text{Fe}^{2+}(3d^6)$ coexist. The amount of Fe^{3+} ions is roughly equal to $4.5 \times 10^{18} \text{ cm}^{-3}$ and the remaining $4.5 \times 10^{19} \text{ cm}^{-3}$ ions are in Fe^{2+} state. This result is represented by middle curve in Fig. 1a. It is worth noticing that the magnetization of this sample is *not* a simple addition of the contributions of $\text{Fe}^{3+}(3d^5)$ and $\text{Fe}^{2+}(3d^6)$ taken separately, as one could ex-

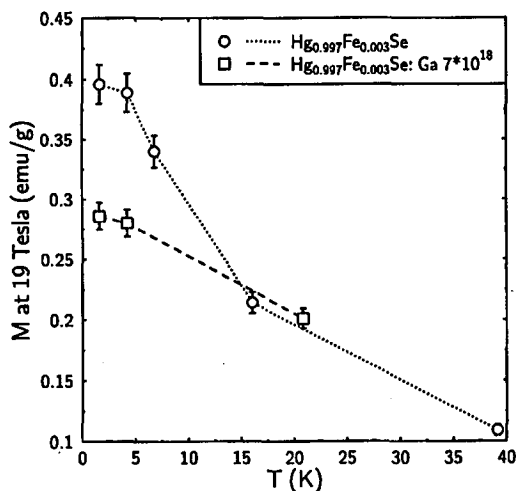


Fig. 2. The absolute value of magnetization taken at 19 T as a function of temperature.

pect. Similar behaviour of magnetization was found by us also for the composition $x = 0.0006$. Moreover, the temperature dependence of the magnetization at 19 T is much stronger than for the sample codoped with gallium (Fig. 2) (containing Fe^{2+} only).

These facts lead to following hypothetical conclusion: there exists an interaction of two magnetic subsystems $\text{Fe}^{3+}(3d^5)$ and $\text{Fe}^{2+}(3d^6)$ which is very sensitive to the number of ions of each species. Of course, this hypothesis needs further theoretical investigations in order to explain the magnetization of diluted HgSe:Fe .

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